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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

This second annual report details progress in the study of optical nonlinearities in molecules at infrared wavelengths. The objectives are to minimize the adverse effects of absorption in both triply resonant (1-, 2-, and 3-photon resonances) and two-photon resonant systems. A second objective is to examine molecules which have unusually strong and narrow Raman active

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modes at the two-photon resonance frequency. The experimental technique is to measure third harmonic generation in the molecular gas using a step tunable CO_2 TEA laser.

Results were obtained for CD_4 at room temperature. The pressure, power, and frequency dependence of the third harmonic signal was measured. Problems were encountered as the fundamental absorption limited the ultimate conversion efficiency. Further experiments are proposed to be conducted at low temperatures which should alleviate this problem.

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NEW NONLINEAR OPTICAL PROCESSES IN MOLECULES AT INFRARED FREQUENCIES

AFOSR-78-3712

I. RESEARCH OBJECTIVES

The overall objective of this research program is to study resonant optical nonlinearities in molecules at infrared wavelengths. The experimental studies all employ third harmonic generation (THG) to probe the nonlinear behavior of different classes of molecules. When used with a step tunable CO_2 laser, this experimental technique can be used to determine the magnitude and spectral dependence of the nonlinear susceptibility.

The first class of optical nonlinearity that has been studied is the purely vibrational nonlinearity which is at least approximately triply resonant. The nearly regular spacing of the molecular energy levels makes simultaneous one-, two-, and three-photon resonances possible. The nonlinear susceptibility in this case is determined exclusively by vibrational transition matrix elements and their detuning factors.

The second class of nonlinearity utilizes only a single two-photon resonance with a vibrational energy level. The susceptibility in this case is determined by electronic transition matrix elements. This susceptibility can also be related to the Raman scattering cross-section. The main factor governing the spectroscopic structure in this case is the detuning from the two photon resonance at the vibrational energy level.

Specific objectives for the current work are the minimization of the adverse effect of absorption in the nonlinear gas at the fundamental laser frequency. For the case of the triple resonance, this necessitates the use of molecules which large rotational constants and

widely spaced vibrational-rotational energy levels. The laser frequency is chosen to lie between one-photon resonances which contribute to absorption. For two-photon resonant molecules, the problem is more simple. Absorptions are due to high J and hot band absorptions from other vibrational minifolds in the molecule. A solution for this problem should be to lower the temperature of the gas and depopulate the absorbing levels.

The second objective of the current work is to enhance the susceptibility of two-photon resonant molecules by choosing species with an unusually narrow and strong two-photon resonance. Cryogenic temperatures will be used in conjunction with this selection process to further concentrate the molecular population in the resonant levels. A further aid to the selection process is the relation between the nonlinear susceptibility for this two-photon resonant case and the spontaneous Raman scattering cross-seciton. Preliminary selection of molecules for study is being done on the basis of a large Raman cross-section.

II. PROGRESS AND ACCOMPLISHMENTS

During the past year, our experimental effort has concentracted on the two-photon resonant molecule, CD_4. An energy level diagram is shown in Fig. 1. It has a strong Raman active resonance at 2108.7 cm^{-1}, which is two-photon resonant with the CO_2 laser line P(12) in the 9 micron band at 1053.024 cm^{-1}. As a symmetric molecule, the two photon resonance of CD_4 has only a single narrow Q-branch. Raman scattering data, Fig. 2, shows this Q-branch to be 15 cm^{-1} in linewidth and to have nearly two times the Raman cross-section as CO, hence one would expect slightly less than four times the THG.

A series of THG experiments have been performed on CD_4 at room temperature, exciting with P(8) to P(16) lines in the 9 micron band of a CO_2 TEA laser. The experimental configuration is shown in Fig. 3. We measured the THG dependence on gas pressure and fundamental laser power for these lines. Due to fundamental absorption at about J=20 on the tail of the ν_4 absorption band, the maximum THG signal occurred at 300 torr pressure. The THG power did have a consistent cubic dependence on fundamental power, as shown for two typical laser lines in Fig. 4. At lower pressures, a factor of three enchancement in THG was observed compared to CO.

The spectral dependence of the THG, shown in Fig. 5, shows an unexpected feature. The 15 cm^{-1} wide ν_1 resonance is expected to be centered at the P(12) line. The increasing trend at the higher frequencies is not predicted by any strong spectral features in the Raman spectrum of CD_4. Unfortunately P(8) is the high frequency limit for lasing in our laser on this band of lines and the behavior at higher

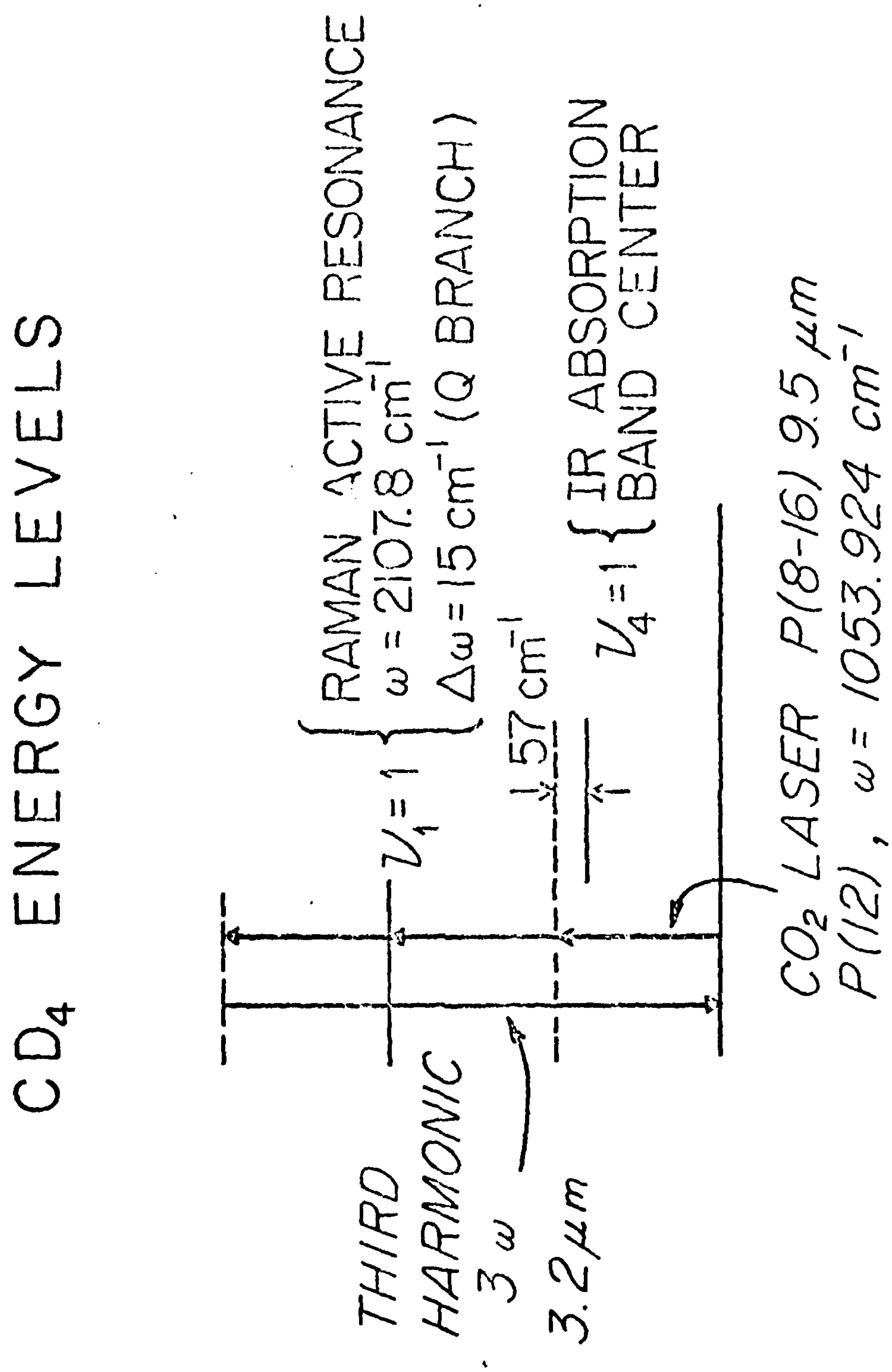
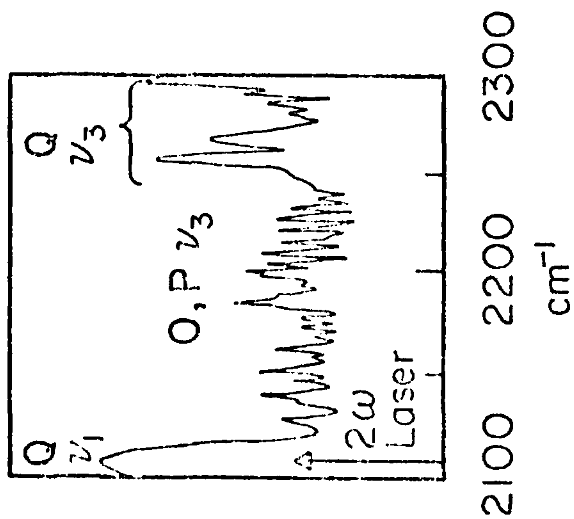

CD_4 ENERGY LEVELS
THIRD HARMONIC
3ω
$3.2\,\mu m$
$\nu_1 = 1$
RAMAN ACTIVE RESONANCE
$\omega = 2107.8\ cm^{-1}$
$\Delta\omega = 15\ cm^{-1}$ (Q BRANCH)
$57\ cm^{-1}$
$\nu_4 = 1$
IR ABSORPTION
BAND CENTER
CO_2 LASER $P(8\text{-}16)$ $9.5\,\mu m$
$P(12)$, $\omega = 1053.924\ cm^{-1}$

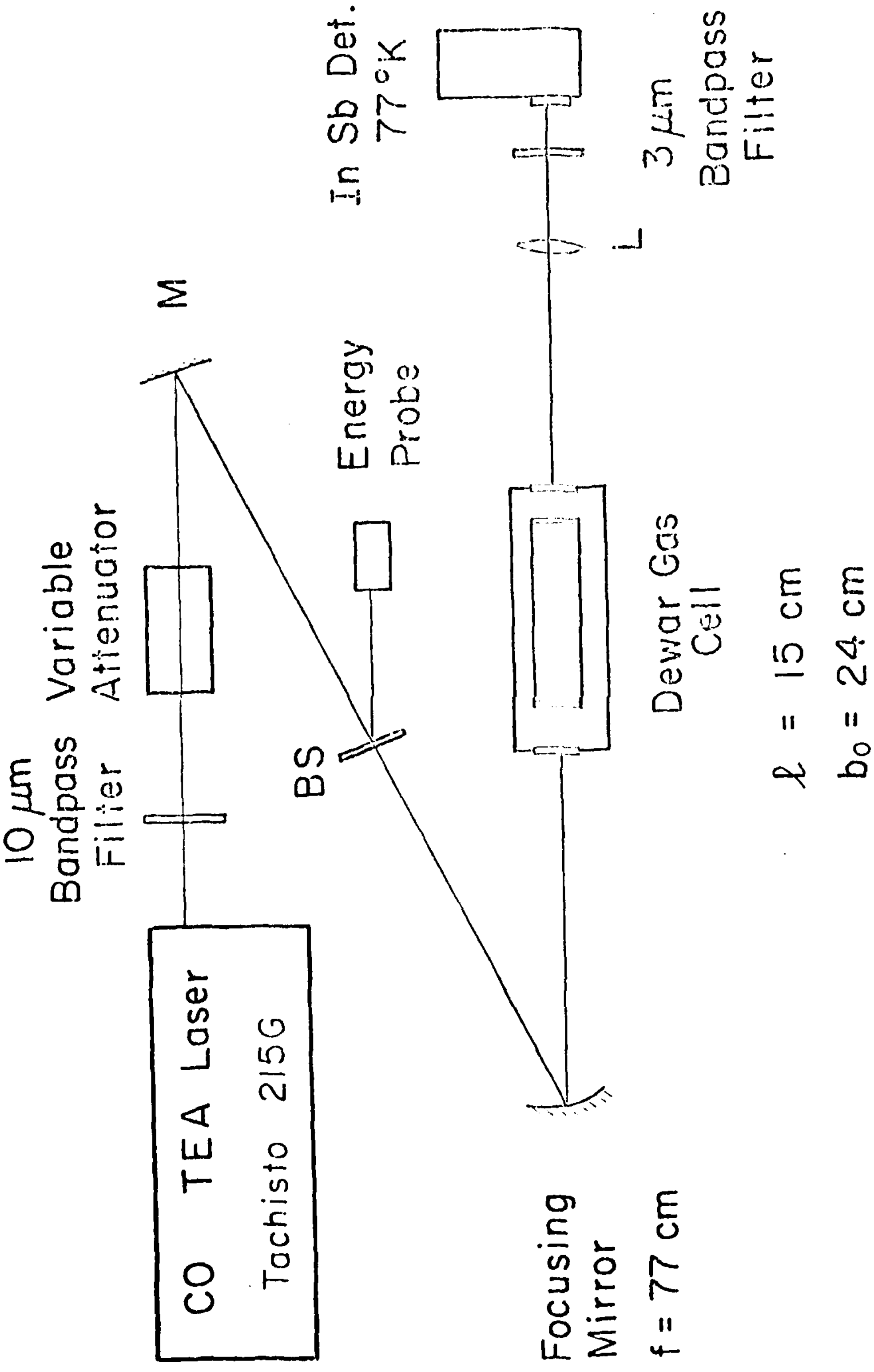
Figure 1



CD_4 RAMAN SPECTRUM

from Sheperd & Welch (1957)

Figure 2



CO TEA Laser
Tachisto 215G
10 μm
Bandpass
Filter
Variable
Attenuator
M
BS
Energy
Probe
Focusing
Mirror
f = 77 cm
Dewar Gas
Cell
ℓ = 15 cm
b₀ = 24 cm
L
3 μm
Bandpass
Filter
In Sb Det.
77°K

Figure 3

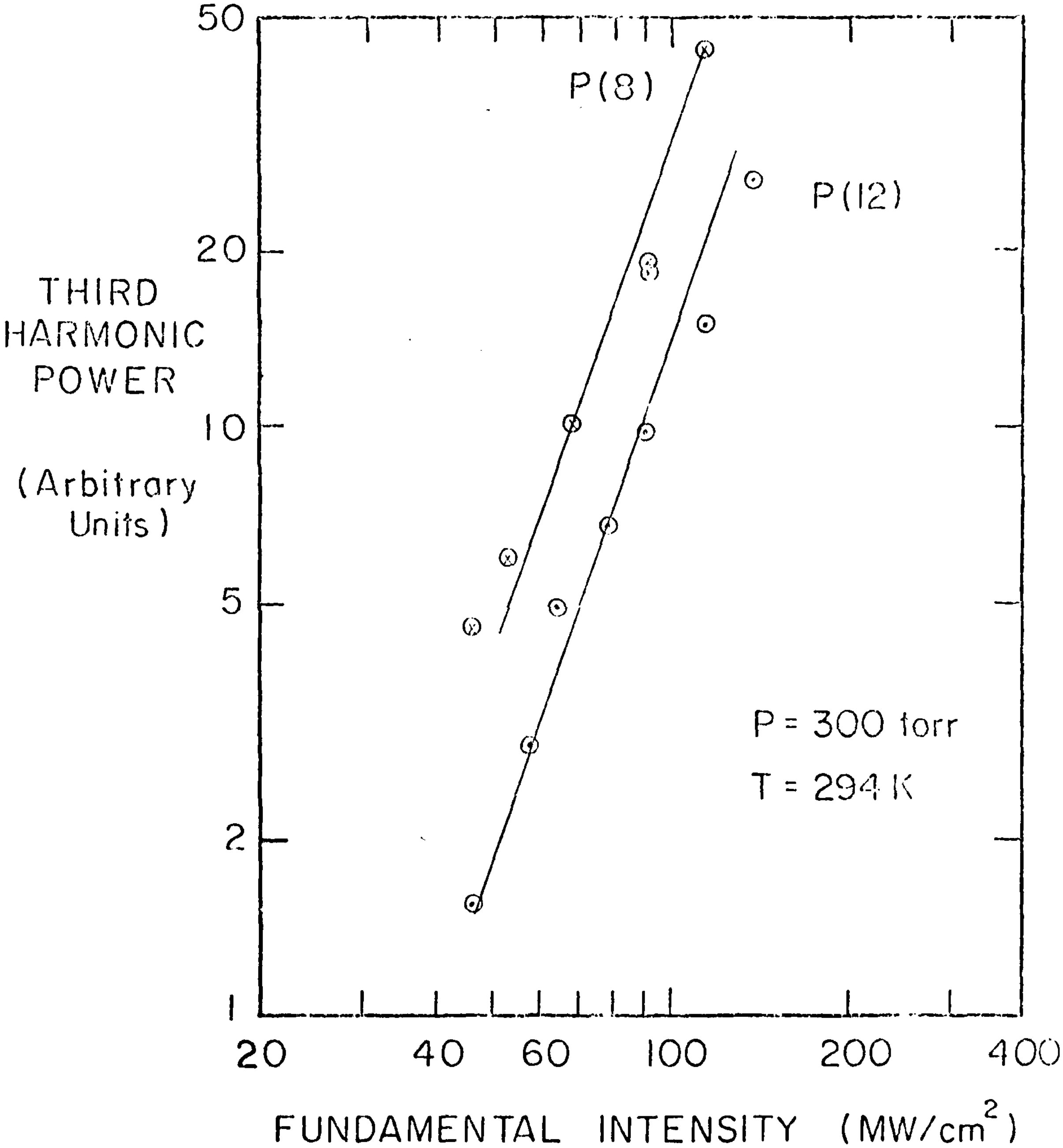

50
20
10
5
2
1
THIRD
HARMONIC
POWER
(Arbitrary
Units)
P(8)
P(12)
P = 300 torr
T = 294 K
20
40
60
100
200
400
FUNDAMENTAL INTENSITY (MW/cm^2)

Figure 4

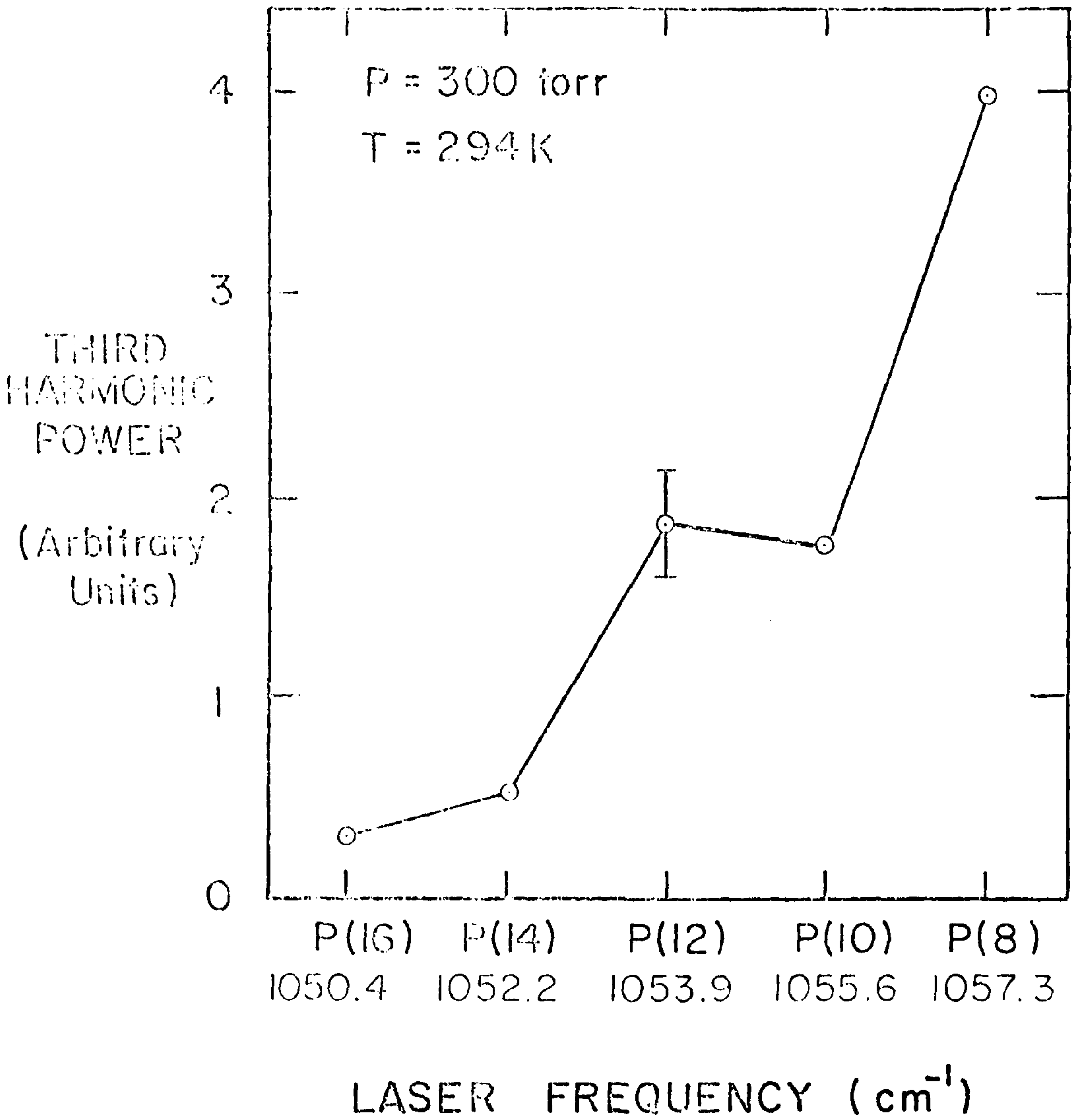

P = 300 torr
T = 294 K
THIRD HARMONIC POWER (Arbitrary Units)
0
1
2
3
4
P(16) 1050.4
P(14) 1052.2
P(12) 1053.9
P(10) 1055.6
P(8) 1057.3
LASER FREQUENCY (cm^{-1})

Figure 5

frequencies could not be studied.

Recent high resolution Raman spectra of CD_4 have been taken by A. Owyoung at Sandia Labs. The data includes part of this region near P(12) but not near P(8). Modeling based on this data is, as a result, incomplete.

These results suggest double benefits to be gained by cooling the gas. First, the high J absorption of the fundamental should decrease dramatically; and second the Raman spectrum in the vicinity of the two-photon resonance should simplify. The result should be higher ultimate conversion efficiency in the first case, and a more easily interpreted THG spectrum, with possibly higher conversion, in the second case.

IV. List of Written Publications

1) K. M. Chung, G. J. Stevens, and M. F. Becker, "Investigation of Multiphoton Absorption in SF_6 by Third Harmonic Generation," IEEE Journal of Quantum Electronics, QE-15, 874, September 1979.

2) M. F. Becker, G. J. Mauger, and Yihjye Twu, "Raman-Resonance Enhanced Third-Harmonic Generation in CD_4," Journal of the Optical Society of America, Vol. 70, 1582, December 1980.

IV. List of Professional Personnel

Dr. Michael F. Becker, Principal Investigator

Mr. G. Joseph Mauger, Graduate Research Assistant

Mr. Yihjye Twu, Graduate Research Assistant

V. Interactions

1. Conference Presentations

M. F. Becker, G. J. Mauger, and Y. Tsu, "Raman-Resonance Enhanced Third Harmonic Generation in CD_4," presented at the Annual Meeting of the Optical Society of America, Chicago, Illinois, October 1980. This conference presentation was highlighted in Laser Focus, pp. 22-24, October 1980.

2. Consultations

June 1980. Visit by M. F. Becker to MIT Lincoln Laboratory to discuss experimental results with Drs. H. Kildal and S. R. J. Brueck.

October 1980. While in Chicago at the Optical Society of America Annual Meeting, met with Drs. P. Esherick and A. Owyoung of Sandia Laboratories to discuss spectroscopic data taken on CD_4.

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